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Brownian drift-diffusion model for evolution of droplet size distributions in turbulent clouds

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[1] Effects from turbulence-induced fluctuations in water vapor saturation on cloud droplet growth are examined using a Brownian diffusion model. The model predicts diffusive broadening of the droplet size distribution, tempered by enhanced evaporation-induced drift of droplets to smaller size from vapor depletion, and approach to a stationary condition (Weibull distribution) determined by the balance between size-space diffusion and drift. Monte Carlo simulations of the approach to the stationary limit and of the distribution itself are presented and compared favorably with observation. A key turbulence parameter required by the kinetic potential theory of drizzle formation is estimated using the new results. **Citation:** McGraw, R., and Y. Liu (2006), Brownian drift-diffusion model for evolution of droplet size distributions in turbulent clouds, *Geophys. Res. Lett.*, *33*, L03802, doi:10.1029/2005GL023545.

1. Introduction

[2] Uncertainty in the physical processes governing clouds and precipitation limits both regional weather forecast accuracy and the ability to predict climate using computer models [*Houghton et al.*, 2001]. A large component of uncertainty is associated with the coupling between cloud turbulence and microphysical processes over a wide range of spatial/temporal scales and droplet size [*Shaw*, 2003]. Current efforts aim at reducing uncertainty through development of more robust parameterizations for clouds and precipitation that are microphysically based yet computationally suitable for use in regional to global scale models [*Rotstayn and Liu*, 2005]. Recent progress in parameterization of clouds and precipitation [*Liu and Daum*, 2000, 2004; *Liu et al.*, 2004, 2005], indirect aerosol effects [*Liu and Daum*, 2002; *Rotstayn and Liu*, 2003; *Peng and Lohmann*, 2003], and rain initiation theory [*McGraw and Liu*, 2003, 2004] reinforces the need for better understanding of the spectral shape of the droplet size distribution.

[3] Although significant progress has been made and a number of models put forth (e.g., stochastic condensation, entrainment and mixing, and systems theory), details of the underlying processes that shape cloud droplet size distributions remain poorly understood [*Baker et al.*, 1980; *Cooper*, 1989; *Srivastava*, 1989; *Khvorostyanov and Curry*, 1999a, 1999b; *Liu et al.*, 2002; *Shaw*, 2003]. Observed droplet size distributions are generally much broader than those predicted by the classical uniform model. Furthermore, few studies/models yield analytical forms for the size distribution that agree well with observations, thus highlighting the need for development of simple microphysics parameterizations [*Liu and Daum*, 2004].

[4] Models of stochastic condensation have usually been of the mean field type. In these a parcel of droplets, estimated on the basis of Kolmogorov scaling to be several meters in extent [*Shaw*, 2003], is uniformly subject to a low-frequency fluctuating saturation tied to the vertical updraft velocity. However, it has been shown that this uniformity places a severe restriction on the degree to which turbulent fluctuations can lead to broadening of the size distribution [*Pruppacher and Klett*, 1997].

[5] Treating fluctuation in the saturation ratio is of itself a long standing problem in cloud physics. A simulation-based approach described by Kulmala and co-workers [*Kulmala et al.*, 1997] captures fluctuations on the smaller spatial scales by sampling the condensation/evaporation trajectories of individual droplets each allowed to experience a different fluctuation history, thus providing a statistical sampling of the droplet distribution. The droplet growth trajectories are assumed to be driven by turbulence fluctuations in vapor saturation. However, effects from vapor depletion (e.g., on slowing of droplet growth and approach to a stationary size distribution) were not included. Here we present a simple model for the effects of turbulence fluctuations based on applying the Langevin and Fokker-Planck equations to the study of cloud droplet size distributions. The model, with vapor depletion included, yields analytic droplet size distributions of the Weibull form in good agreement with observation.

2. Turbulent Condensation and Evaporation: Diffusive Growth of Cloud Droplets

[6] Turbulence causes fluctuations in water vapor saturation and, consequently, in the rate of droplet growth. Such fluctuations play an essential role in broadening of the cloud droplet size distribution and can be modeled either by Monte Carlo simulation [*Kulmala et al.*, 1997] or analytically, as described below, in terms of a Fokker-Planck equation describing the drift and diffusion of droplets along a coordinate of droplet size.

[7] Cloud droplet growth/evaporation takes place in the continuum regime for which the rate is:

$$\frac{dr^2}{dt} = k(T)(S-1) = k(T)(\langle S\rangle - 1) + k(T)(S(t) - \langle S\rangle) \quad (1)$$

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where r is droplet radius, and $k(T)$ is a temperature and pressure dependent rate coefficient (to simplify notation we suppress the weaker pressure dependence) that includes coupled heat and mass transfer during growth/evaporation of the drop [*Pruppacher and Klett*, 1997]. S is the saturation ratio, defined as the ratio of the vapor pressure of the interstitial cloud air to the equilibrium vapor pressure of the drop. We assume that nearby droplets within a parcel are subject to the same value of $S(t)$ due to rapid diffusion of water vapor among the droplets. For diffusive equilibration times of $10s \approx \gamma^{-1}$ (see below) vapor concentrations will be uniform over distances of order 1 cm [*Clement*, 2003] while droplets that are further apart experience different fluctuation histories. The second equality of equation (1) allows for the possibility that the average parcel saturation ratio $\langle S \rangle$ may be other than unity as a consequence of adiabatic cooling or vapor depletion (see below). We assume fluctuations in S characterized by finite variance σ_S^2,

$$\left\langle (S - \langle S \rangle)^2 \right\rangle = \sigma_S^2, \tag{2a}$$

with exponential decay of correlation over timescale γ^{-1},

$$\begin{aligned} \langle (S(t) - \langle S \rangle)(S(t+\Delta) - \langle S \rangle) \rangle &= \langle S(t)S(t+\Delta) - \langle S \rangle^2 \\ &= \langle S(0)S(\Delta) \rangle - \langle S \rangle^2 \\ &= \sigma_S^2 \exp(-\gamma\Delta) \end{aligned} \tag{2b}$$

This agrees with the model of Kulmala and co-workers [*Kulmala et al.*, 1997], however it is significant that we will not require the fluctuations in S to be gaussian. Thus the present analysis can apply even in the face of large non-gaussian fluctuations in S from intermittency – a well known property of cloud turbulence [*Shaw*, 2003]. Estimates for σ_S (on the order of 1%) and for the correlation time, γ^{-1} (from several seconds to tens of seconds) are available [*Kulmala et al.*, 1997]. These short correlation times suggest that fluctuations in S are strongly damped over the time scale, τ, estimated below, of significant change in the droplet size distribution. Finally it is assumed (c.f. the second equality of equation (2b)) that the fluctuations are stationary in the sense that their statistical properties depend only on the time difference, Δ.

[8] The preceding suggests an analogy to Brownian particle motion with coordinate $z \equiv r^2$ and equations (1) and (2a) (2b) giving the instantaneous velocity, $v = dz/dt$. The latter has two components: the fluctuation term in equation (1), containing $S(t)$ and giving rise to diffusion and, as we will show that $\langle S \rangle < 1$, a depletion term, proportional to $\langle S \rangle - 1$, giving rise to drift. The full problem, including both diffusion and drift, is analogous to the well-studied model of Brownian motion in a field of force, allowing one to write down many key results immediately rather than having to repeat in detail derivations available in standard texts [e.g., *Serra et al.*, 1986; *Gardiner*, 1985]. Consider first the random growth component. In the strongly damped regime ($\gamma^{-1} \ll \tau$) this causes droplets to diffuse along the z coordinate with diffusion coefficient given by the product of the variance of the growth velocity fluctuations, $k^2(T)\sigma_S^2$, and correlation time [*Serra et al.*, 1986]:

$$D_Z = \frac{k^2(T)\sigma_S^2}{\gamma}. \tag{3}$$

3. Vapor Depletion and the Stationary Cloud Droplet Distribution

[9] Diffusion of droplet size is checked by requirements that the droplet radius be positive and total water (liquid plus vapor) be conserved; droplets cannot grow without vapor depletion, which will be represented here in a mean field sense by assigning an averaged saturation $\langle S \rangle$ to the cloud parcel under consideration. This average is expected to change slowly with reference to the correlation time scale γ^{-1} as the distribution changes (section 4), and approach an asymptotic value under stationary cloud conditions, which can be determined self-consistently by the methods now described.

[10] For $\langle S \rangle \neq 1$ the first term on the right hand side of equation (1) gives a deterministic drift in droplet size with velocity:

$$v_{depl} = \left(\frac{dr^2}{dt} \right)_{depl} = k(T)(\langle S \rangle - 1). \tag{4}$$

Just as in the case of Brownian motion, the combination of diffusion and drift is described by a Fokker-Planck (FP) equation [*Serra et al.*, 1986]. In present notation:

$$\frac{\partial f}{\partial t} = D_Z \frac{\partial^2 f}{\partial z^2} - v_{depl} \frac{\partial f}{\partial z} = \frac{k^2(T)\sigma_S^2}{\gamma} \frac{\partial^2 f}{\partial z^2} - v_{depl} \frac{\partial f}{\partial z}. \tag{5}$$

(A similar FP equation, but of widely different scale, has been used in nucleation theory to describe the drift and diffusion of molecular clusters along the coordinate of cluster size [*Lifshitz and Pitaevskii*, 1981]). The stationary condition ($\partial f/\partial t = 0$) is determined by the balance between diffusion, which tends to broaden the distribution, and increase liquid water content, and drift, which tends to narrow the distribution, and decrease liquid water content, by reducing droplets to smaller size (v_{depl} must be negative for a stationary distribution). Equation (5) yields a Boltzmann distribution in z as its stationary solution:

$$f_\infty(z) = N_D \frac{|v_{depl}|}{D_Z} \exp\left(-\frac{|v_{depl}|}{D_Z} z \right), \tag{6}$$

where $|v_{depl}| = -v_{depl}$ is the magnitude of v_{depl}, normalization is to the droplet number concentration N_D, and the subscript on f refers to the stationary condition. The liquid water fraction (cm^3 cloud liquid water/cm^3 air) is obtained as the 3/2 moment of $f(z)$:

$$L = \frac{4\pi}{3} \int_0^\infty z^{3/2} f(z) dz. \tag{7}$$

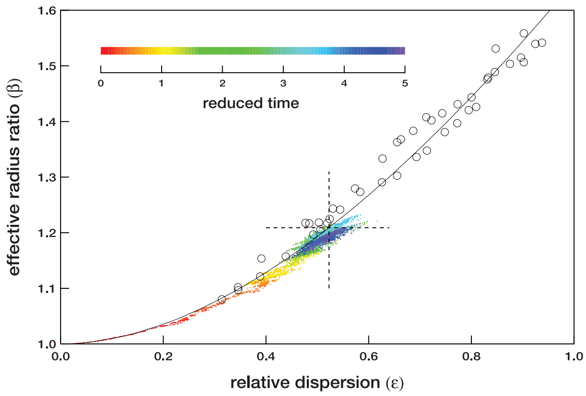

Figure 1. Color points, relative dispersion, ε, and effective radius ratio, β, for an evolving 100-drop sample taken at reduced time increments of 0.001 (5000 samples total) as a function of reduced sample time (see legend). Results are shown for evolution from an initially monodisperse size distribution (red) to the stationary distribution (purple). Fluctuations are due to the stochastic nature of the model and limited sample size. Open circles, results from measurements of droplet size distributions in marine and continental clouds [see *Liu et al.*, 2002]. Solid curve, analytic results for family of Weibull distributions. Dashed line segment intersection, $(\varepsilon_\infty, \beta_\infty)$ for the stationary distribution from the present theory (equation (10)).

Substitution of $f_\infty(z)$ for specified liquid water content yields the stationary value of v_{depl}:

$$v_{depl} = -\pi\left(\frac{N_D}{L}\right)^{2/3} D_Z = -\pi\left(\frac{N_D}{L}\right)^{2/3}\frac{k^2(T)\sigma_S^2}{\gamma}. \qquad (8)$$

Reflective of vapor depletion, the theory predicts a uniform shift in droplet averaged saturation to values below unity. From equations (4) and (8):

$$\langle S\rangle_\infty = 1 - \pi\left(\frac{N_D}{L}\right)^{2/3}\frac{k(T)\sigma_S^2}{\gamma}, \qquad (9)$$

and the stationary average saturation can thus be determined from the parameter estimates for σ_S and γ provided in section 5. In the absence of fluctuations ($\sigma_S^2 = 0$) water vapor is in equilibrium with the droplets and $S = 1$.

[11] Transforming equation (6) from z to droplet radius gives the Weibull distribution:

$$f_\infty(r) = 2\pi N_D\left(\frac{N_D}{L}\right)^{2/3} r \exp\left[-\pi\left(\frac{N_D}{L}\right)^{2/3} r^2\right], \qquad (10)$$

which is a good representation of typically observed cloud droplet size distributions [*Costa et al.*, 2000; *Liu et al.*, 2002] (see Figure 1).

4. Monte Carlo Simulation

[12] For simulation we replace equation (5) with the equivalent Langevin equation:

$$dz = v_{depl}dt + \sigma_Z dX \qquad (11)$$

where $\sigma_Z^2 = 2D_Z$ and $dX = \phi\sqrt{dt}$. ϕ is a dimensionless random variable drawn from a standardized normal distribution with zero mean and unit variance, $p(\phi) = (2\pi)^{-1/2}\exp(-\phi^2/2)$. With these definitions, $\langle dX\rangle = 0$ and $\langle dX^2\rangle = dt$. Equivalence of equations (5) and (11) is demonstrated in standard texts on stochastic processes [e.g., *Gardiner*, 1985]. The drift-diffusion processes they describe are frequently encountered and well suited to simulation using Monte Carlo methods. Simulations are carried out here for *100*-drop samples of growth/evaporation trajectories based on equation (11). Droplets interact through the vapor depletion effect, but are otherwise independent as they are assumed far enough apart to experience different fluctuation histories. At each time step the drift velocity is adjusted so as to preserve liquid water content close to its specified value $L(t)$. Generally, e.g., with a parcel undergoing adiabatic cooling, L will be a function of time and N_D will change with the activation to new droplets or droplet loss. To illustrate the new methods we here assume the simplest case of fixed values for L and N_D specified by the initial condition. Time is expressed in units of the distribution relaxation time mentioned above, $\tau = z_0^2/(2D_Z)$, where $z_0 = (3/4\pi)^{2/3}(L/N_D)^{2/3}$ is the average radius squared of the droplets, and radius in units of $r_0 = \sqrt{z_0}$. Scaled results are thus independent of L, N_D and D_Z. The model time step is set at $0.001\tau(d\tilde{t} = 0.001)$ and simulations carried out to $t = 5\tau(\tilde{t} = 5)$. Positive values for the size coordinate are assured by applying a reflective boundary condition at the origin.

[13] Figure 1 shows evolution of the relative dispersion, ε, defined as the square root of the variance of the droplet radial distribution divided by its mean, and effective radius ratio, β, defined as the ratio of the third to second radial moments divided by the cube root of the third moment [*Liu et al.*, 2002]. The color points show the values of $\varepsilon(\tilde{t}_k)$ and $\beta(\tilde{t}_k)$ at each successive time step, $\tilde{t}_k = 0.001k$, over the course of the simulation. The initial distribution is taken to be monodisperse ($\varepsilon(0) = 0$), broadening with time due to the turbulence fluctuations in saturation and growth rates, maintaining constant L and N_D. Note that the broadening seen here is counter to the usual tendency of condensation growth at fixed (non-fluctuating) saturation to narrow size distributions over time [*McGraw*, 1997]. Broadening is effectively complete by $\tilde{t} = 1-2$ with slower approach to the asymptotic values, $\varepsilon_\infty = \sqrt{4/\pi - 1} = 0.5227\ldots$, $\beta_\infty = (9\pi/2)^{1/3}/2 =$

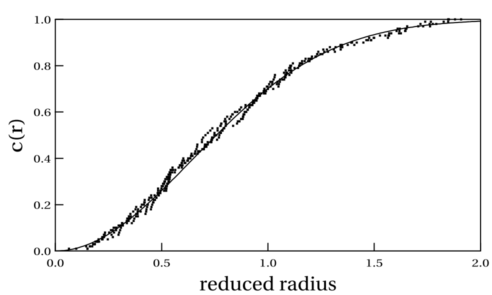

Figure 2. Cumulative radial distribution versus scaled drop radius from equation (10) (solid curve) and comparison with results from four 100-drop Monte-Carlo simulations (400 points total) at different times near the stationary limit (purple point region of Figure 1).

1.20899..., marked by intersection of the dashed line segments in Figure 1, as predicted from the radial moments of the stationary distribution, equation (10). Figure 2 shows the cumulative radial distribution (normalized to unity) for the Weibull distribution (equation (10)) and comparison with results combining four 100-drop Monte Carlo simulations at different times near the stationary limit.

5. Implications for the Study of Clouds and Precipitation

[14] The simple Brownian drift-diffusion model, using only the continuum growth law (equation (1)) and a reasonable model for fluctuations in S (equations (2a) and (2b)), has been found to yield a Weibull distribution of cloud droplet size in good agreement with observations. Nevertheless the model does not reproduce the broader measured droplet spectra seen in Figure 1. This may be a limitation of using fixed values for L/N_D in the simulations rather than averaging over a range of values for this parameter as likely to be found in turbulent clouds. A wider family of distributions can be obtained by allowing for drift/diffusion coefficients that are functions of droplet size. For example, the Kelvin effect, not included here, gives a small additional, size dependent contribution to the drift to smaller droplet size. Alternatively, empirical size distributions can be used and equation (5) inverted to obtain the drift/diffusion rates. Extensions of the method to allow for size-dependent drift/diffusion rates and variable liquid water fraction will be described elsewhere.

[15] The present analysis provides a key turbulence parameter used in the kinetic potential (KP) theory of drizzle formation [*McGraw and Liu*, 2003, 2004]. This is the quantity $t_{1\%}$, defined as the time required for diffusion along the growth coordinate to change the cloud droplet size 1% from 10 to 10.1 micron radius. This can now be expressed in terms of the diffusion constant: $t_{1\%} = (\Delta z)^2/(2D_Z)$, where $\Delta z = 10.1^2 - 10.0^2 = 2.01\ \mu\mathrm{m}^2$ and the units of D_Z are $\mu\mathrm{m}^4\ \mathrm{s}^{-1}$. We estimate D_Z from equation (3): Using $k(10°\mathrm{C}) = 167.8\ \mu\mathrm{m}^2\ \mathrm{s}^{-1}$, from equation (13.28) and the parameters given in Table 13.1 of *Pruppacher and Klett* [1997], saturation variance $\sigma_S = 0.01$ and correlation time $\gamma^{-1} = 7\mathrm{s}$, both from *Kulmala et al.* [1997], yields $D_Z = 20.2\ \mu\mathrm{m}^4\ \mathrm{s}^{-1}$ and $t_{1\%} = 0.1\mathrm{s}$. This is in the range of previous very rough estimates for this parameter and happens to be a condition for which detailed calculations of the drizzle barrier and drizzle rate have been presented [*McGraw and Liu*, 2003, 2004]. For a typical mean droplet radius $r_0 = 10\ \mu\mathrm{m}$ ($z_0 = 100\ \mu\mathrm{m}^2$) we obtain the estimate $\tau = z_0^2/(2D_Z) \approx 4$ min for the distribution relaxation time, thereby justifying the strongly damped condition ($\gamma^{-1} \ll \tau$) used in derivation of the Fokker-Planck and equivalent Langevin equations. In conclusion, the new theory provides both a mechanism for shaping the cloud droplet distribution and foundation for the similar drift-diffusion processes that underlie the KP theory of drizzle initiation.

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